



CASE STUDY I:

Age determination of uranium materials

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Radioactivity, Radionuclides & Radiation
Nuclides.net training course



Nuclear Forensic Science

- Aim is the identification of unknown nuclear materials
- Motivated through increasing number of cases of illicit trafficking involving nuclear materials (approx. 500 confirmed cases during the last ten years)
- e.g. 1994: Munich airport seizure: 560 g mixed PuO_2 , U_3O_8 + 210 g ^6Li (89.4% abund.)

Questions:

- nature of material?
- danger associated?
- offence committed?
- origin, possible route?
 - intended use?



Material identification

- Visual inspection, weight, photography
- Isotopic composition: HRGS, TIMS, SIMS
- Concentration of main components: Titration, IDMS
- Impurities: ICP-MS
- Material properties: TEM (internal microstructure, lattice defects)
SEM (grain size, surface structure)
- Date of production (“Age”): ICP-MS, TIMS, Alpha-Spectrometry



Principle of age determination

- “Age” (or production date) refers to the date of the last separation of the parent nuclide from its daughters
- Age is calculated from the mass ratio or activity ratio of mother nuclide over daughter nuclide grown in
- Assumes complete removal of daughter nuclides during the initial purification process
- Assumes absence of subsequent contamination



The age of uranium material is calculated from the basic equation of radioactive decay

$$N = N_0 \times e^{-\lambda t}$$

An example for age calculation from the $^{234}\text{U}/^{230}\text{Th}$ ratio:

$$\frac{N_{U-234}}{N_{Th-230}} = \frac{N_{0U-234} \times e^{-\lambda_{U-234}t}}{(N_{0U-234} - N_{U-234}) \times e^{-\lambda_{Th-230}t}}$$

time or "age" t can be solved from:

$$t = \frac{\ln\left(1 - \frac{R}{K}\right)}{\beta}$$

where R is the **measured $^{230}\text{Th}/^{234}\text{U}$ -atom ratio**

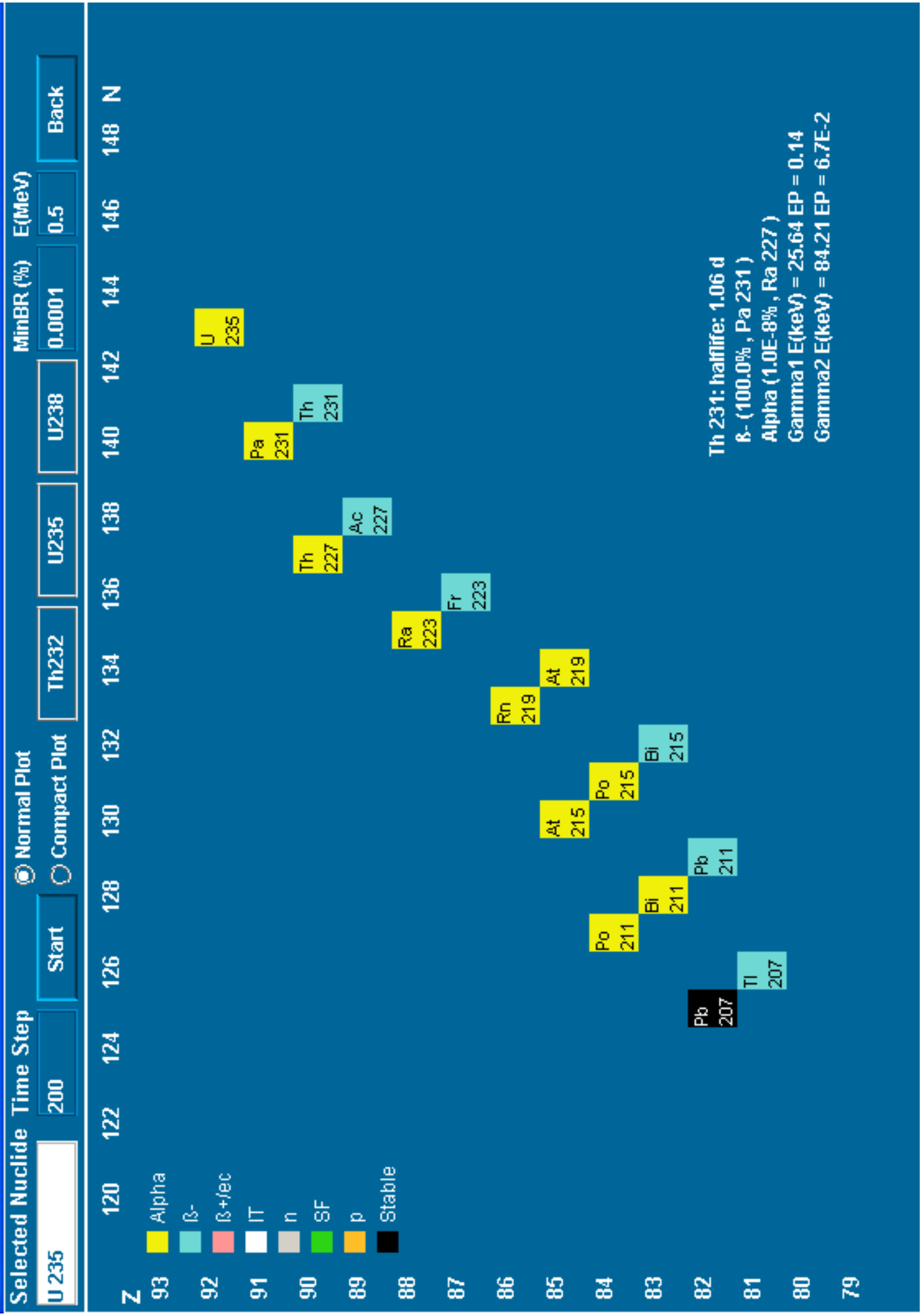
β is a factor composed of the ^{234}U and ^{230}Th decay constants = $(\lambda_{U-234} - \lambda_{Th-230})$

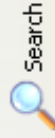
$$K = \frac{\lambda_{U-234}}{(\lambda_{Th-230} - \lambda_{U-234})}$$



Isotopic compositions of typical uranium materials:

	U-234 (Wt.%)	U-235 (Wt.%)	U-236 (Wt.%)	U-238 (Wt.%)
Natural uranium	0.005	0.711	-	99.284
Low enriched U-030	0.002	3.006	0.021	96.955
High enriched U-850	0.641	84.987	0.372	14.000





Address http://141.52.159.201/Nuclides/Decay/N2K_FD_Engine.asp?fr=VB&uk=01742207031038010001&mi=922340

Go Links



234 U

Full Decay

Options

Quantity:

Grams



1.0000E-03

Time:

Years



20

Distance(cm):

100

Number of time steps:

1

Min. Prod.:

1E-02

N° Chains:

1

Start

Reset

Parent+Daughters	N(atoms)	M(g)	A(Bq)	G(keV/s)	dH/dt(μSv/hr)
92 U234	2.5730E+18	9.9994E-04	2.3020E+05	2.4685E+04	8.3885E-03
90 Th230	1.4518E+14	5.5457E-08	4.2304E+01	1.5697E+01	1.0772E-06
88 Ra226	1.3303E+10	4.9929E-12	1.8262E-01	1.1178E+00	6.8283E-10
86 Rn222	8.6905E+04	3.2039E-17	1.8235E-01	6.5097E-02	1.0318E-11
84 Po218	4.8931E+01	1.7714E-20	1.8235E-01	1.6785E-03	2.4623E-13
82 Pb214	4.2293E+02	1.5029E-19	1.8231E-01	4.1345E+01	1.7803E-08
83 Bi214	3.1404E+02	1.1159E-19	1.8231E-01	2.8019E+02	3.7540E-08
84 Po214	0.0000E+00	0.0000E+00	0.0000E+00	0.0000E+00	0.0000E+00
82 Pb210	3.3038E+07	1.1520E-14	3.2541E-02	6.8091E-02	3.7566E-09
83 Bi210	2.0276E+04	7.0700E-18	3.2449E-02	9.3789E-06	2.9444E-15
84 Po210	5.1802E+05	1.8063E-16	3.0033E-02	2.6531E-04	3.9236E-14
82 Pb206 Stable	4.7418E+06	1.6218E-15	0.0000E+00	0.0000E+00	0.0000E+00
Total :	2.5731E+18	1.0000E-03	2.3024E+05	2.5024E+04	8.3896E-03

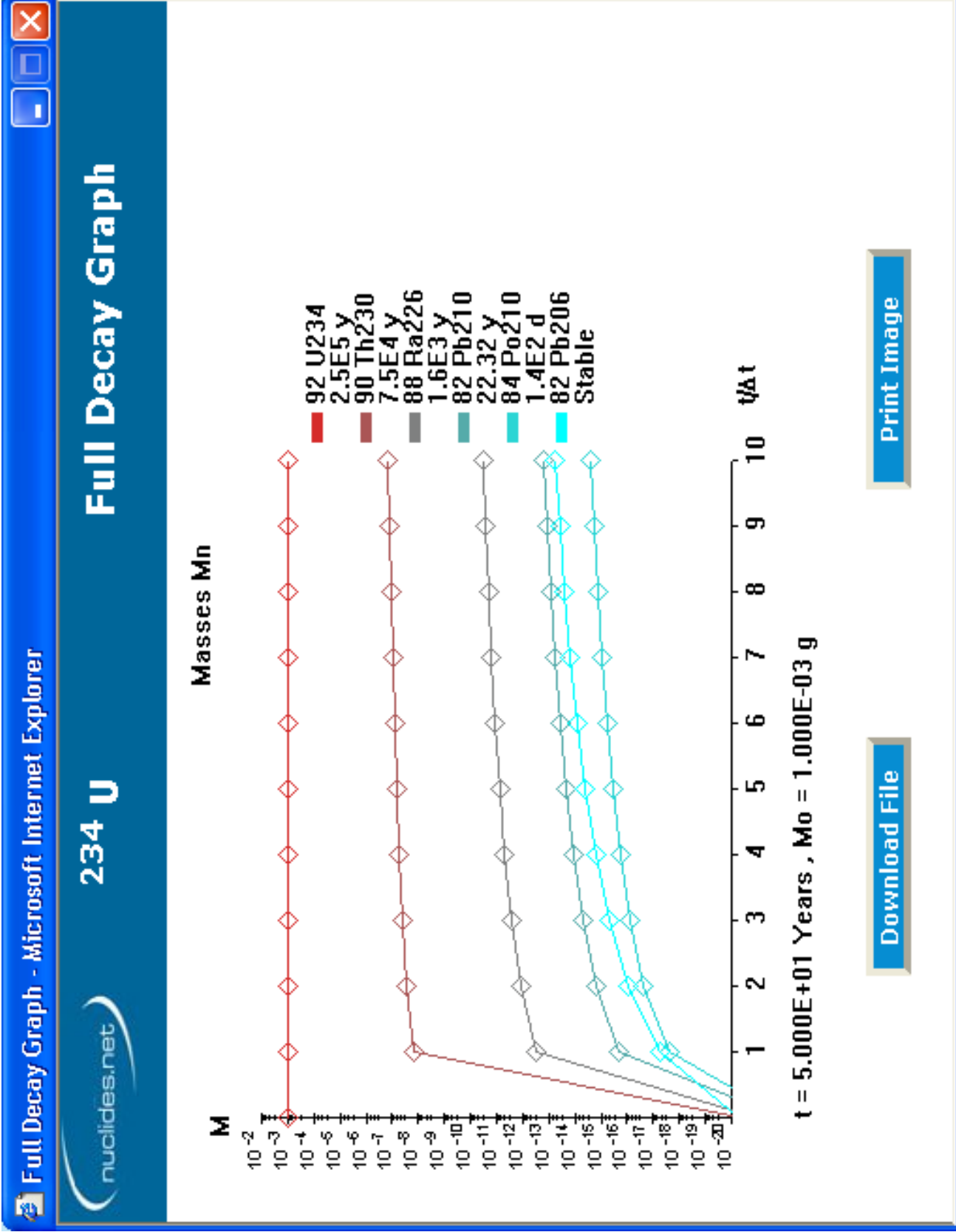
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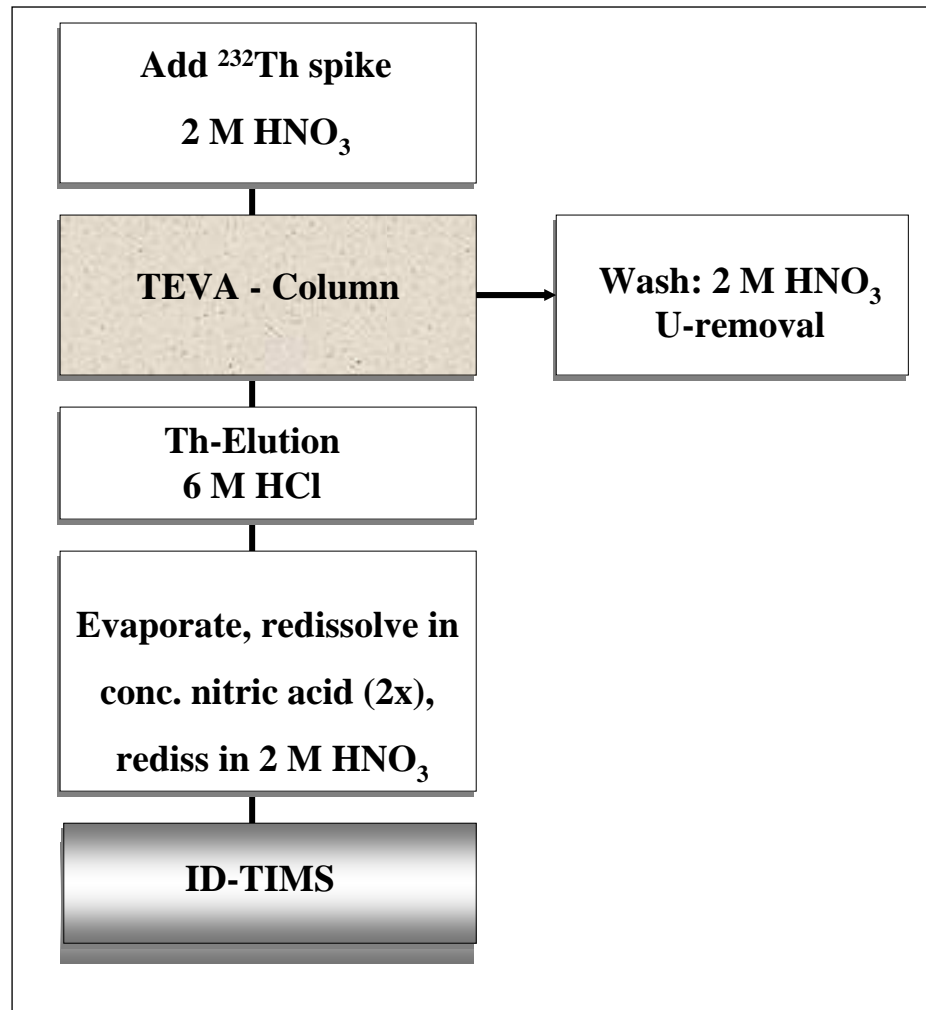


Parent+Daughters	N(atoms)	M(g)	A(Bq)
92 U234	2.5730E+18	9.9994E-04	2.3020E+05
90 Th230	1.4518E+14	5.5457E-08	4.2304E+01
88 Ra226	1.3303E+10	4.9929E-12	1.8262E-01
86 Rn222	8.6905E+04	3.2039E-17	1.8235E-01
84 Po218	4.8931E+01	1.7714E-20	1.8235E-01
82 Pb214	4.2293E+02	1.5029E-19	1.8231E-01
83 Bi214	3.1404E+02	1.1159E-19	1.8231E-01
84 Po214	0.0000E+00	0.0000E+00	0.0000E+00
82 Pb210	3.3038E+07	1.1520E-14	3.2541E-02
83 Bi210	2.0276E+04	7.0700E-18	3.2449E-02
84 Po210	5.1802E+05	1.8063E-16	3.0033E-02
82 Pb206 Stable	4.7418E+06	1.6218E-15	0.0000E+00
Total :	2.5731E+18	1.0000E-03	2.3024E+05





Analysis of $^{234}\text{U}/^{230}\text{Th}$ via ID-TIMS

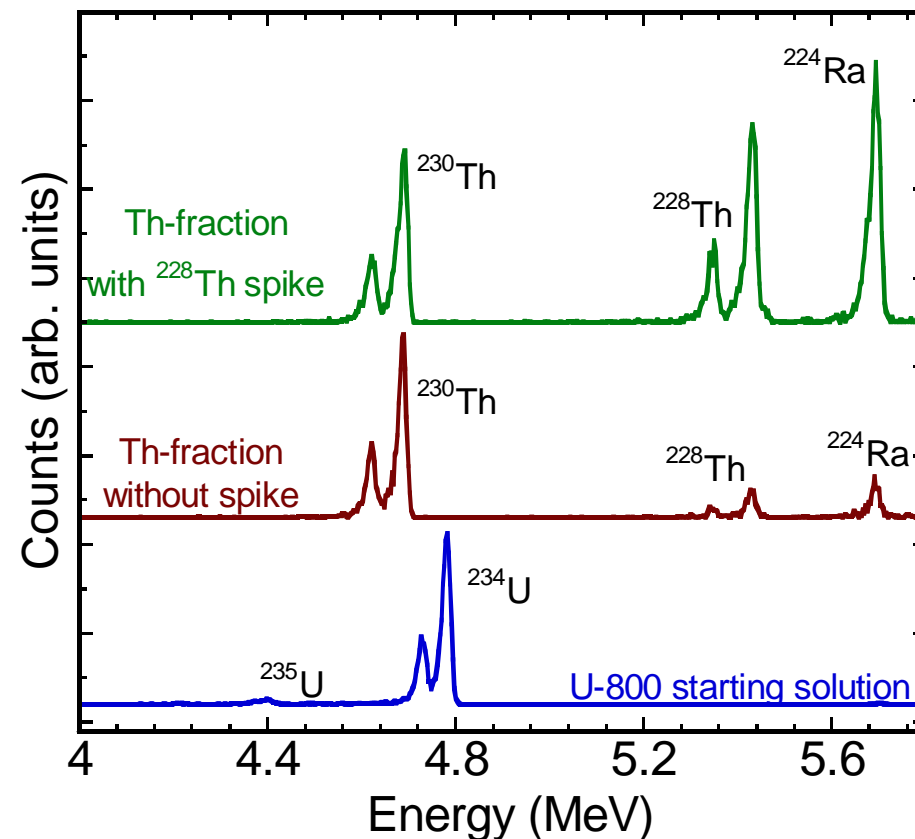
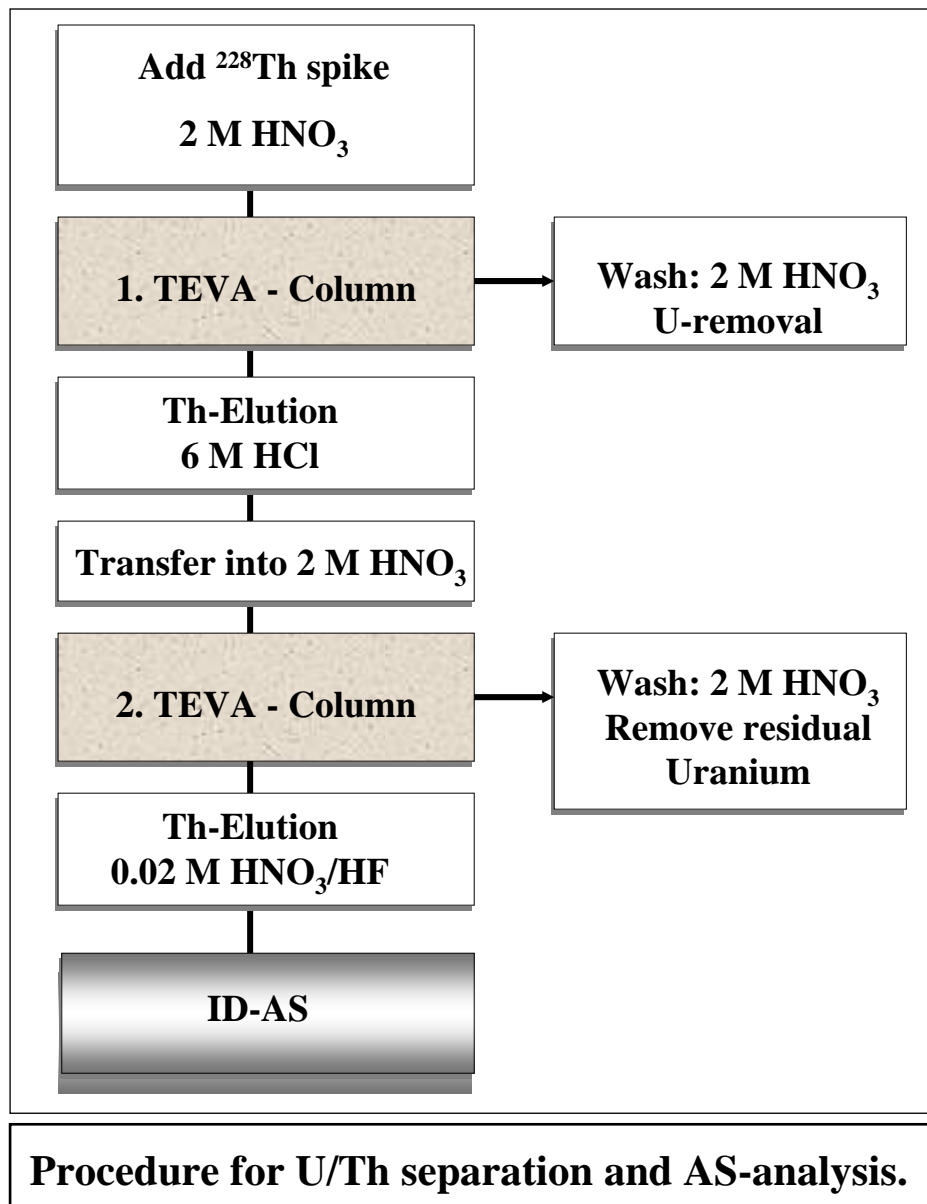


Extraction chromatographic procedure for U/Th separation and TIMS-analysis.

Sample (^{235}U -abund.)	Assumed Age (y)	Determined Age (y)
U-100 (10%)	43.7	44.0±1.3
U-500 (50%)	39.1	38.8±0.4
U-800 (80%)	43.3	42.7±0.6
U-850 (85%)	44.0	43.3±0.8



Analysis of $^{234}\text{U}/^{230}\text{Th}$ via ID-AS



Alpha-spectra of U solution and Th-fractions (spiked and unspiked). The U spectrum corresponds to $1.5 \mu\text{g U}_3\text{O}_8$, the Th-spectra were measured from $6.3 \text{ ng } ^{230}\text{Th}$ extracted from $4.2 \text{ mg U}_3\text{O}_8$.

Address http://141.52.159.201/Nuclides/Decay/N2K_FD_Engine.asp?fr=VB&uk=01742207031038010001&mi=922350**235 U**

Full Decay

Options

Quantity:

Grams



1E-3

Time:

Years



30

Distance(cm):

100

Number of time steps:

1

Min. Prod.:

1E-02

N° Chains:

2

Start**Reset**

Parent+Daughters	N(atoms)	M(g)	A(Bq)	G(keV/s)	dH/dt(μSv/hr)
92 U235	2.5621E+18	1.0000E-03	7.9960E+01	1.1955E+04	1.5043E-05
90 Th231	1.0598E+07	4.0659E-15	7.9960E+01	1.0522E+03	2.3586E-05
91 Pa231	7.5614E+10	2.9009E-11	5.0697E-02	1.5948E+00	1.0468E-08
89 Ac227	1.7873E+07	6.7381E-15	1.8031E-02	3.1582E-03	6.3907E-10
90 Th227	4.1316E+04	1.5576E-17	1.7706E-02	1.7580E+00	2.7449E-09
88 Ra223	2.5525E+04	9.4529E-18	1.7908E-02	1.5455E+00	2.6955E-09
86 Rn219	0.0000E+00	0.0000E+00	0.0000E+00	0.0000E+00	0.0000E+00
84 Po215	0.0000E+00	0.0000E+00	0.0000E+00	0.0000E+00	0.0000E+00
82 Pb211	5.5960E+01	1.9606E-20	1.7908E-02	1.1966E+00	2.1848E-10
83 Bi211	3.3173E+00	1.1622E-21	1.7908E-02	8.1491E-01	2.1813E-10
81 Tl207	7.3738E+00	2.5343E-21	1.7859E-02	3.8984E-02	5.7580E-12
82 Pb207 Stable	6.0236E+06	2.0703E-15	0.0000E+00	0.0000E+00	0.0000E+00
87 Fr223	0.0000E+00	0.0000E+00	0.0000E+00	0.0000E+00	0.0000E+00
Total :	2.5621E+18	1.0000E-03	1.6008E+02	1.3014E+04	3.8646E-05

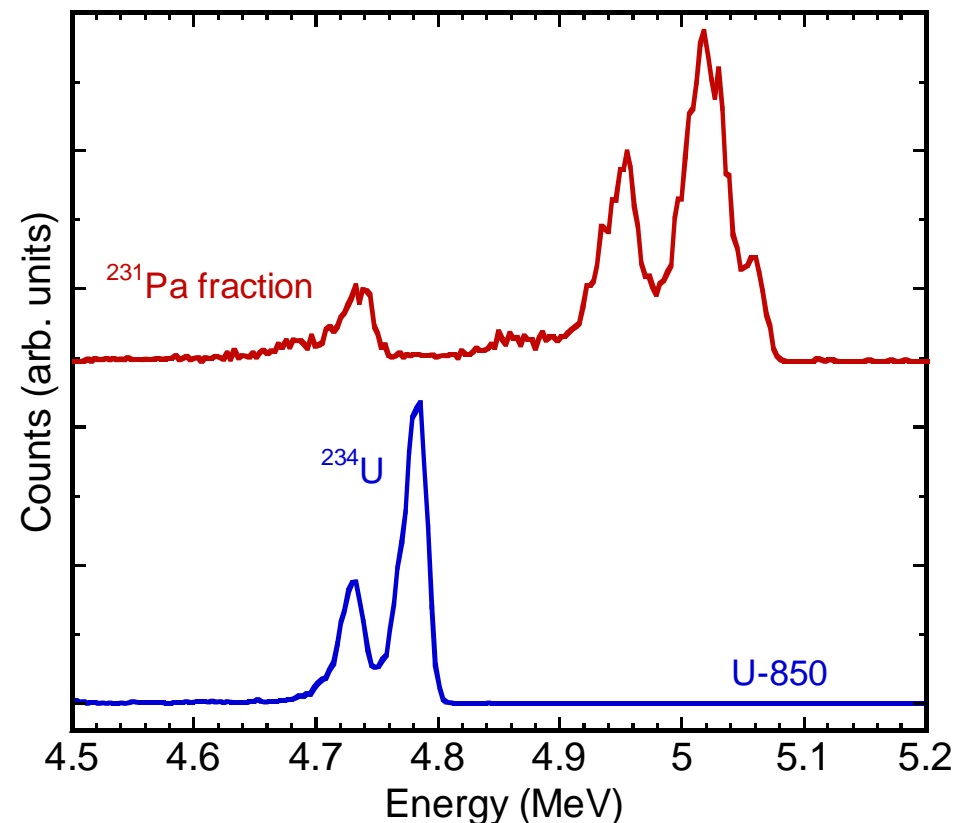
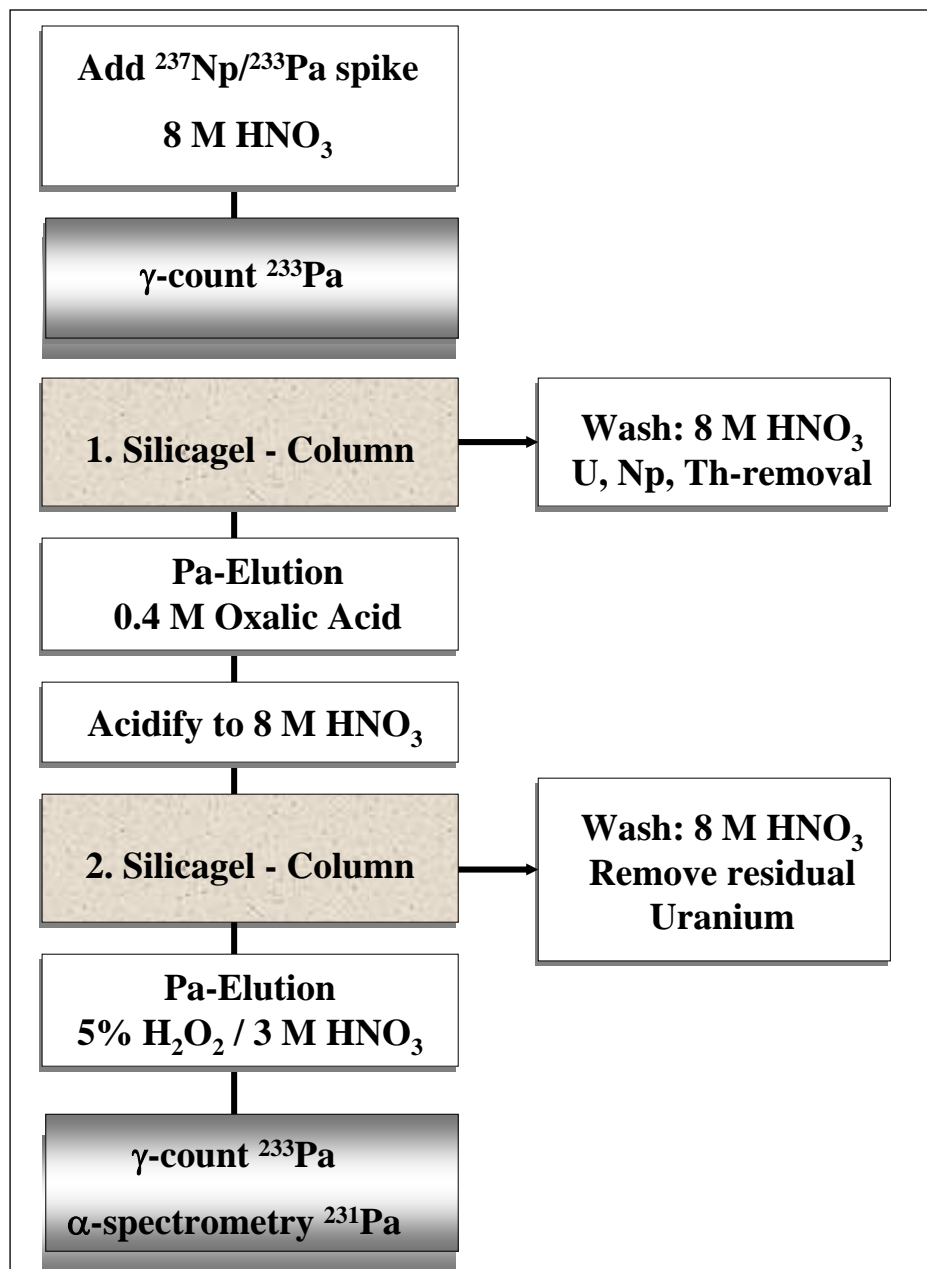
Print**Graph****Details**



Parent+Daughters	N(atoms)	M(g)	A(Bq)
92 U235	2.5621E+18	1.0000E-03	7.9960E+01
90 Th231	1.0598E+07	4.0659E-15	7.9960E+01
91 Pa231	7.5614E+10	2.9009E-11	5.0697E-02
89 Ac227	1.7873E+07	6.7381E-15	1.8031E-02
90 Th227	4.1316E+04	1.5576E-17	1.7706E-02
88 Ra223	2.5525E+04	9.4529E-18	1.7908E-02
86 Rn219	0.0000E+00	0.0000E+00	0.0000E+00
84 Po215	0.0000E+00	0.0000E+00	0.0000E+00
82 Pb211	5.5960E+01	1.9606E-20	1.7908E-02
83 Bi211	3.3173E+00	1.1622E-21	1.7908E-02
81 Tl207	7.3738E+00	2.5343E-21	1.7859E-02
82 Pb207 Stable	6.0236E+06	2.0703E-15	0.0000E+00
87 Fr223	0.0000E+00	0.0000E+00	0.0000E+00
Total :	2.5621E+18	1.0000E-03	1.6008E+02



Analysis of $^{235}\text{U}/^{231}\text{Pa}$ via ID-AS

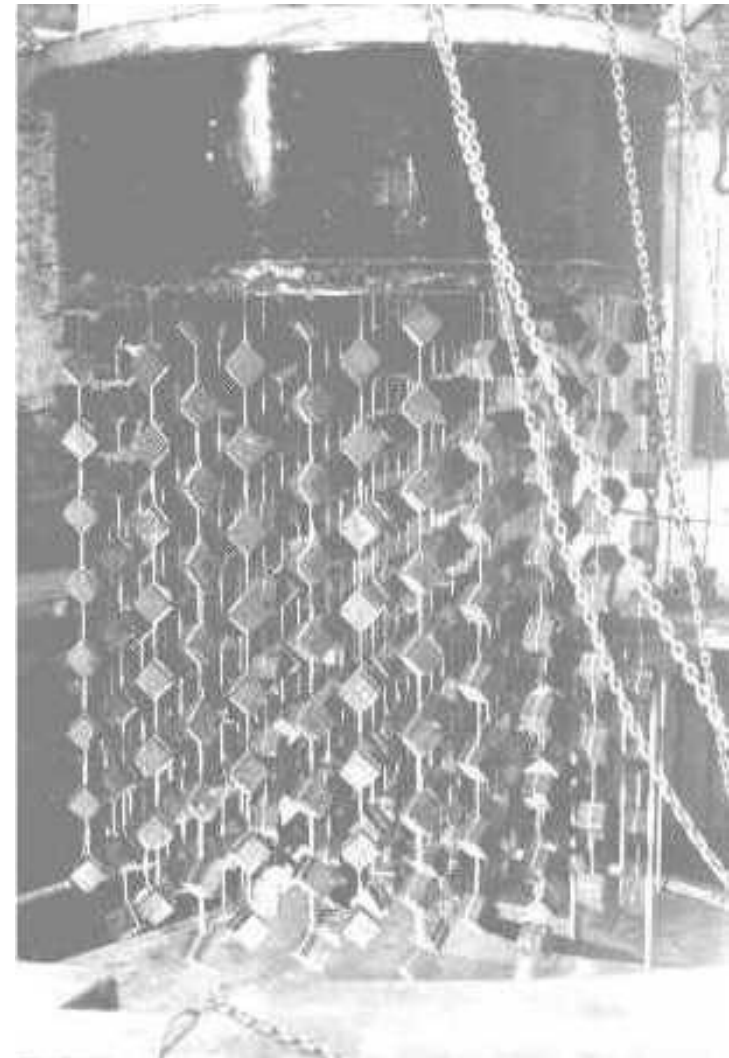


Alpha-spectra of U-850 solution and extracted Pa-fraction. The U spectrum corresponds to $1.5 \mu\text{g U}_3\text{O}_8$, the spectrum of Pa was measured from $150 \text{ pg } ^{231}\text{Pa}$ extracted from $10.2 \text{ mg U}_3\text{O}_8$.



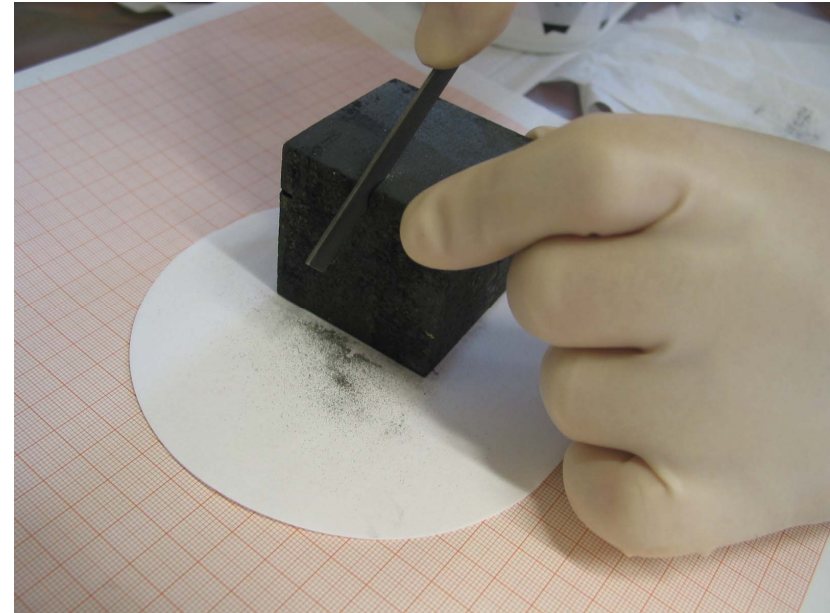
Werner Heisenberg's uranium machine

- During WWII German scientists led by Werner Heisenberg studied the feasibility of a nuclear chain reaction
- 1944/45 experimental setup located in a cave in Haigerloch, Germany





- Reactor consisted of 664 cubes of uranium metal, 2.4 kg each
- moderator D_2O
- setup never went critical for lack of materials and time
- 659 cubes seized by American troops
- 5 cubes disappeared

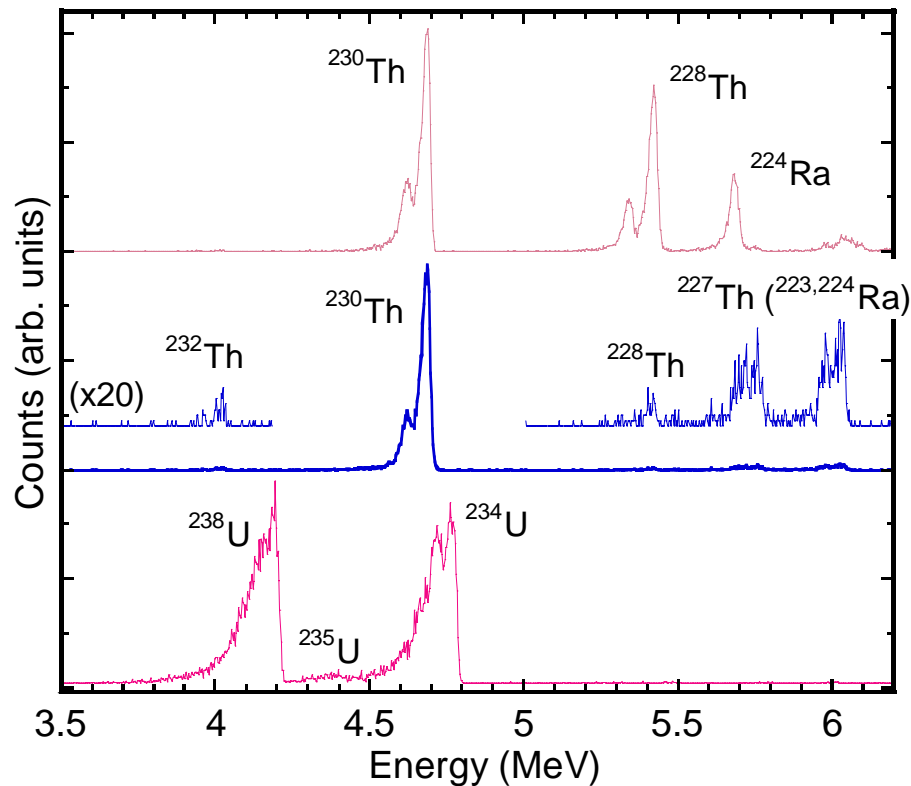




U - Isotopic composition (TIMS)

U-234	U-235	U-238
54 ppm	0.72 %	99.28 %

Age via ID-AS ^{234}U / ^{230}Th



Exercise:

Mass ratio Th-230 / U-234:

$1.636\text{e-}4$

=> Find the age using

Nuclides Net “Full decay”

Options

Quantity:

Grams

Time:

Years




Distance(cm):

Min. Prod.:

Number of time steps:

N° Chains:

Parent+Daughters	N(atoms)	M(g)	A(Bq)	G(keV/s)	dH/dt(μSv/hr)
92 U234	2.5726E+21	9.9982E-01	2.3017E+08	2.4682E+07	8.3874E+00
90 Th230	4.7171E+17	1.8019E-04	1.3745E+05	5.1002E+04	3.4998E-03
88 Ra226	1.3958E+14	5.2388E-08	1.9161E+03	1.1729E+04	7.1645E-06
86 Rn222	9.1280E+08	3.3652E-13	1.9153E+03	6.8375E+02	1.0838E-07
84 Po218	5.1394E+05	1.8605E-16	1.9153E+03	1.7630E+01	2.5863E-09
82 Pb214	4.4422E+06	1.5786E-15	1.9149E+03	4.3426E+05	1.8699E-04
83 Bi214	3.2985E+06	1.1721E-15	1.9149E+03	2.9430E+06	3.9431E-04
84 Po214	0.0000E+00	0.0000E+00	0.0000E+00	0.0000E+00	0.0000E+00
82 Pb210	8.4667E+11	2.9522E-10	8.3394E+02	1.7450E+03	9.6273E-05
83 Bi210	5.2068E+08	1.8155E-13	8.3327E+02	2.4085E-01	7.5612E-11
84 Po210	1.4060E+10	4.9026E-12	8.1515E+02	7.2012E+00	1.0649E-09
82 Pb206 Stable	4.4966E+11	1.5380E-10	0.0000E+00	0.0000E+00	0.0000E+00
Total :	2.5731E+21	1.0000E+00	2.3032E+08	2.8124E+07	8.3916E+00

Address  http://141.52.159.201/Nuclides/Decay/N2K_FD_Engine.asp?fr=VB&uk=01742207031038010001&mi=922340 Go  Links

234 U

Full Decay

Options

Quantity:

Grams

▼

1

Time:

Years

▼

59

Distance(cm):

100

Min. Prod.:

1E-02

Number of time steps:

1

N° Chains:

1

Start

Reset

Parent+Daughters	N(atoms)	M(g)	A(Bq)	G(keV/s)	dH/dt(μSv/hr)
92 U234	2.5727E+21	9.9983E-01	2.3017E+08	2.4683E+07	8.3875E+00
90 Th230	4.2819E+17	1.6356E-04	1.2477E+05	4.6296E+04	3.1769E-03
88 Ra226	1.1510E+14	4.3201E-08	1.5801E+03	9.6721E+03	5.9081E-06
86 Rn222	7.5269E+08	2.7749E-13	1.5793E+03	5.6382E+02	8.9369E-08
84 Po218	4.2380E+05	1.5342E-16	1.5793E+03	1.4538E+01	2.1326E-09
82 Pb214	3.6630E+06	1.3017E-15	1.5790E+03	3.5809E+05	1.5419E-04
83 Bi214	2.7199E+06	9.6654E-16	1.5790E+03	2.4268E+06	3.2514E-04
84 Po214	0.0000E+00	0.0000E+00	0.0000E+00	0.0000E+00	0.0000E+00
82 Pb210	6.5633E+11	2.2885E-10	6.4646E+02	1.3527E+03	7.4630E-05
83 Bi210	4.0359E+08	1.4073E-13	6.4588E+02	1.8669E-01	5.8608E-11
84 Po210	1.0871E+10	3.7907E-12	6.3028E+02	5.5680E+00	8.2342E-10
82 Pb206 Stable	3.1335E+11	1.0717E-10	0.0000E+00	0.0000E+00	0.0000E+00
Total :	2.5731E+21	1.0000E+00	2.3031E+08	2.7525E+07	8.3913E+00

Print

Graph

Details



Result:

Age: 59.0 ± 1.5 years

=> Date of production: December 1943

=> Sample seems authentic